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# Pulsed Power for a Dynamic Transmission Electron Microscope

w. j. dehope, n.d. browning, g.h. campbell, e.g. cook,  
w.e. king, t.b. lagrange, b.w. reed, b.c. stuart, R.M.  
Shuttlesworth, B.J. Pyke

June 29, 2009

Pulsed Power for a Dynamic Transmission Electron  
Microscope

washington , DC, United States

June 29, 2009 through July 2, 2009

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# Pulsed Power for a Dynamic Transmission Electron Microscope

W.J. DeHope, N.D. Browning, G.H. Campbell,  
E.G. Cook, W.E. King, T.B. LaGrange, B.W. Reed,  
B.C. Stuart, R.M. Shuttlesworth, B.J. Pyke

*Lawrence Livermore National Laboratory*

**17th IEEE International Pulsed Power Conference  
Washington , DC, United States  
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# Image gathering is an old science



- 200 um spatial resolution
- 15 ms temporal resolution
- dynamic range  
100 (inst.)  
100k (long-term)
- BW=400-700nm
- ease-of-focus/use
- advanced image  
processing/correction
- 28k “pixels”
- redundancy
- long life





# Early primitive optical microscopes ushered in modern science



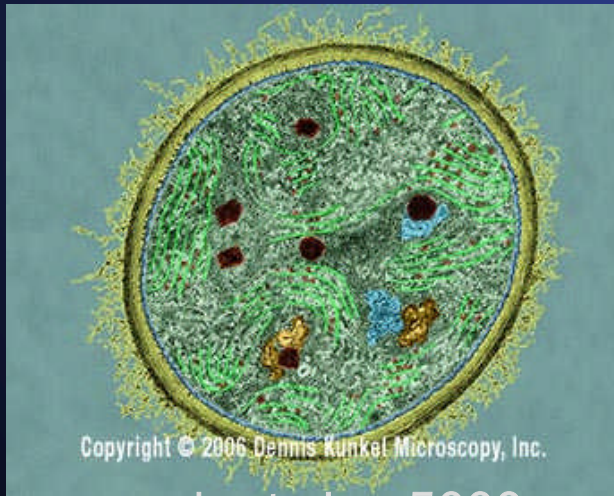
- Robert Hooke publishes *Micrographia* in 1665
- Replica of Leeuwenhoek's 1675 instrument (300X with single lens)
- SoA 1780
- Commercial product ~1850



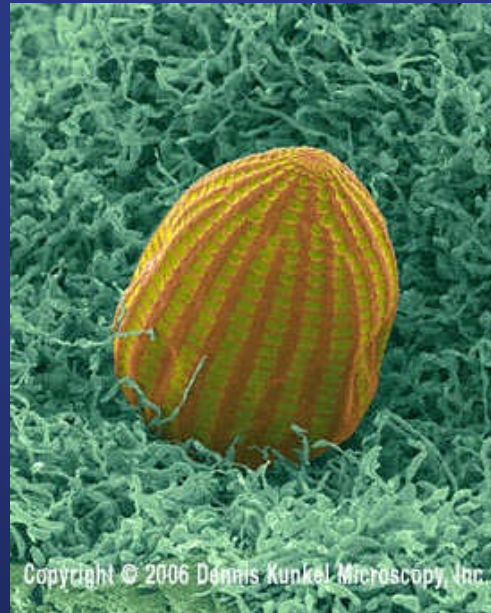
# Remember your first microscope?



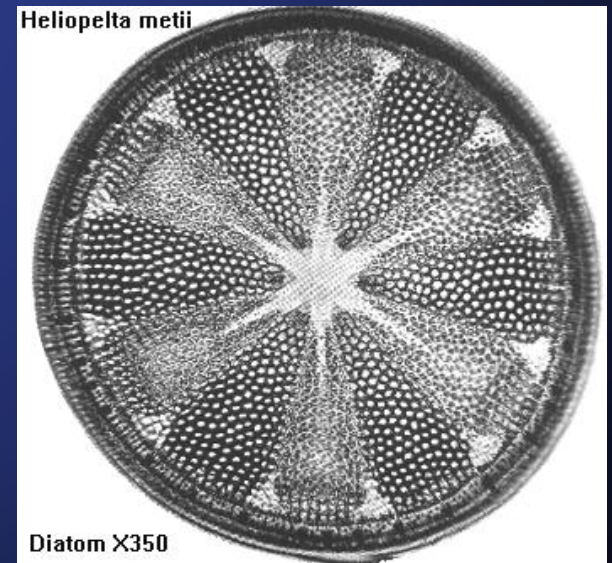
Ushered many of us into science



cyanobacterium 5000x

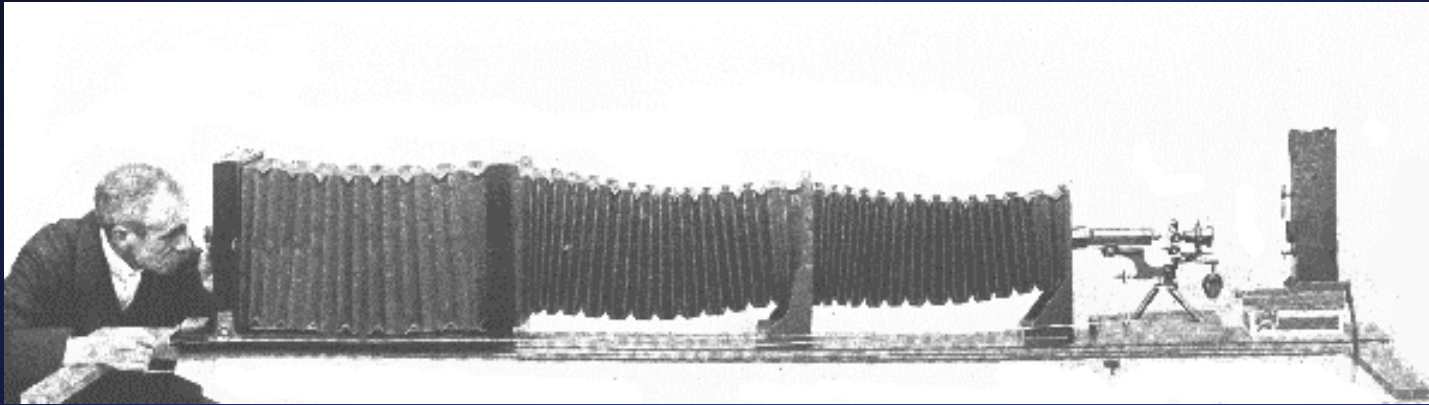


monarch butterfly egg 10x



diatom 350X (1904)

# 1% inspiration, 99%...



*Nature through Microscope and Camera*, Kerr & Smith, 1904

- resolution limit = wavelength / N.A.
- ~300nm
- ~500X
- Optical microscopy was a mature technology by the 20<sup>th</sup> cent.
- We need something shorter than the wavelength of light!
- deBroglie wavelength of electron:  $h/p$   
 $2.4/\beta\gamma$  pm

*Micrographia*, Hooke, 1664

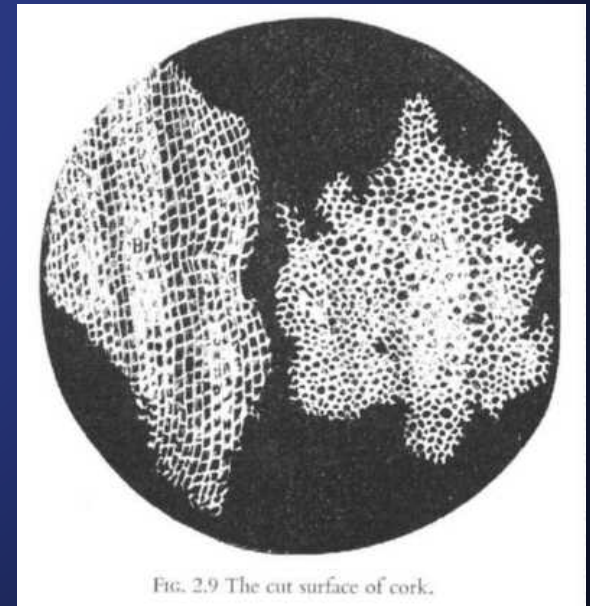
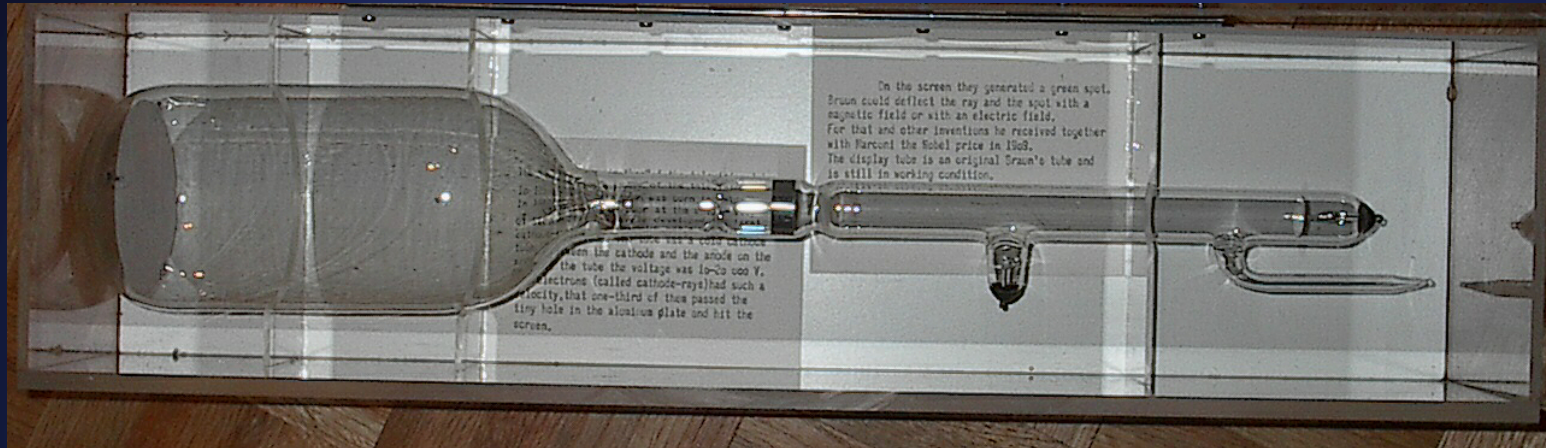


FIG. 2.9 The cut surface of cork.



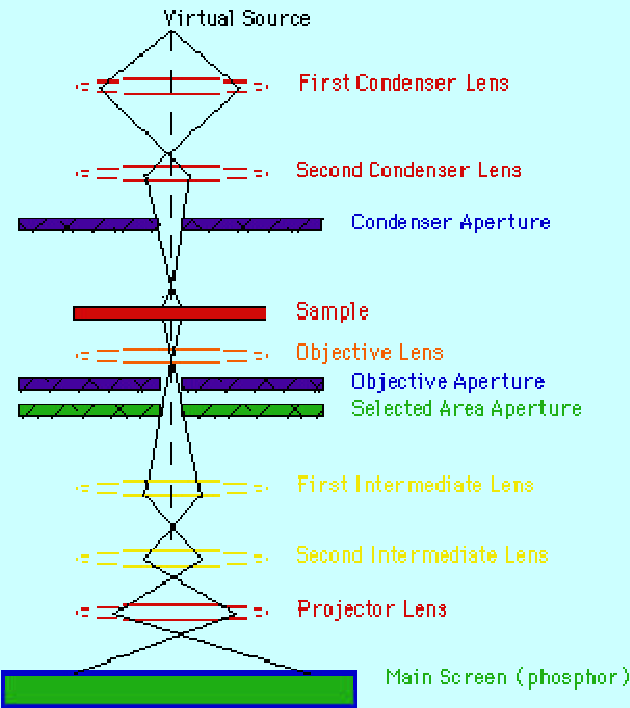
# Early electron tubes saw advances in vacuum and electron optics



- Edison light bulb 1880
- Braun CRT 1897
- Fleming diode 1904
- deForest ad 1929
- Varian klystron 1938
- Kompfner TWT 1943

# Transmission Electron Microscope

## Max Knoll & Ernst Ruska 1932



- Eli Burton, U Toronto 1938
- RCA 1939
- JEOL 1947
- CW images
  - 0.1nm
  - 50M X
- temporal res'n: 1 ms



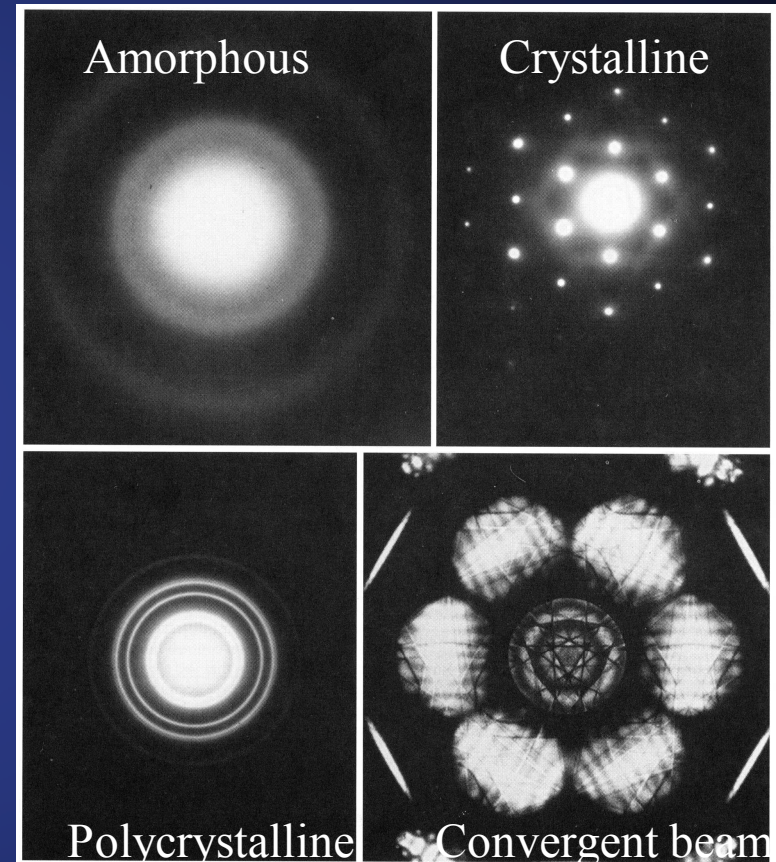
1/29/2009



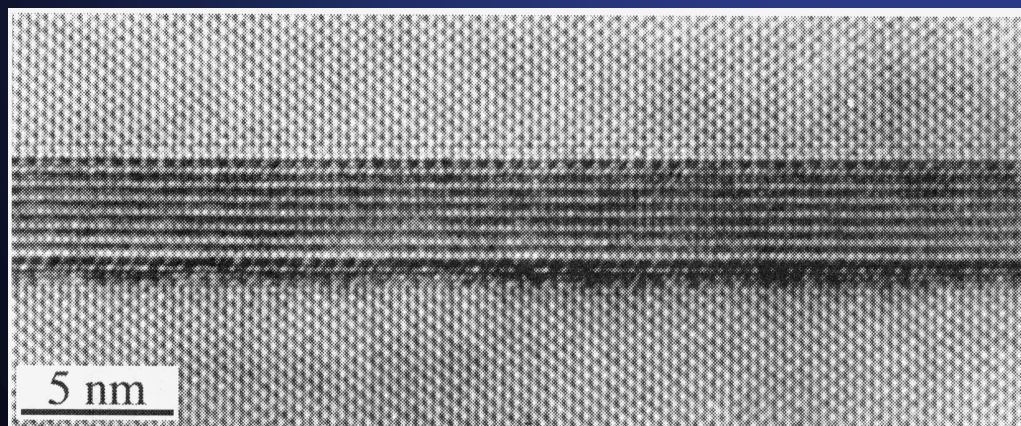
# The strengths of TEM



- TEM excels at directly measuring the *structure* of a material
  - Shape, from nm to mm scales
  - Defects and strain
  - Atomic arrangements on 0.1 nm scale
  - Crystal structure (or lack thereof)
  - Which elements are where, and how they're bonded to each other



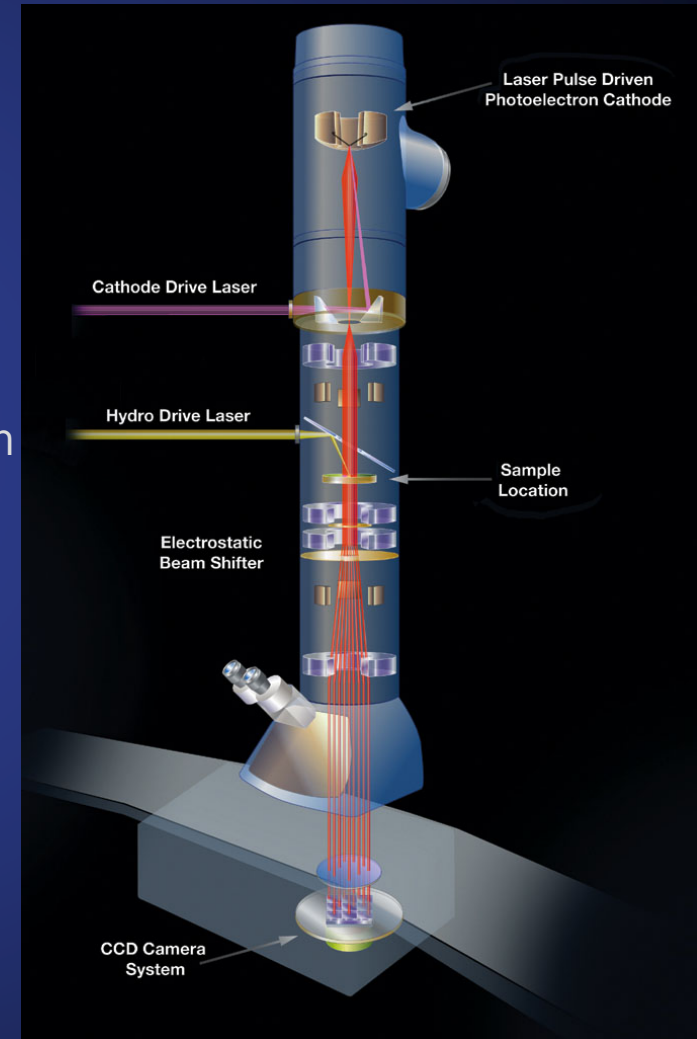
Figures from D. B. Williams & C. B. Carter, Transmission Electron Microscopy, Plenum, 1996.





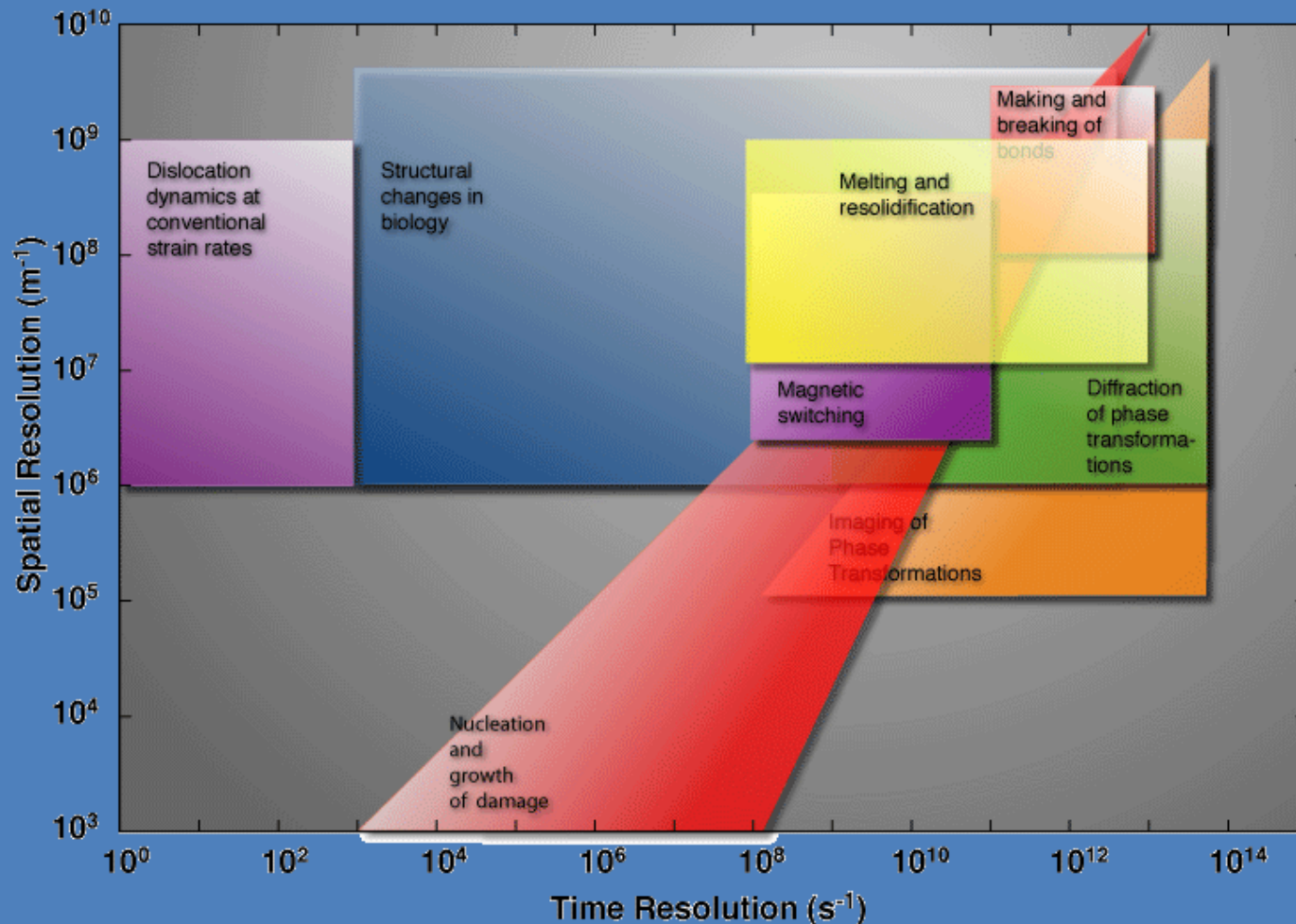
# Motivation for this work

- How do we maintain the spatial resolution of TEM for short pulses?
- Can we image material phase transitions?
- Can we separately shock materials as image response is measured?
- Can we image molecular processes on relevant time scales?
- Such a device would be a  
Dynamic  
Transmission  
Electron  
Microscope  
or DTEM!





# High temporal resolution opens up fundamental processes in Biology, Chemistry, & Matl Science



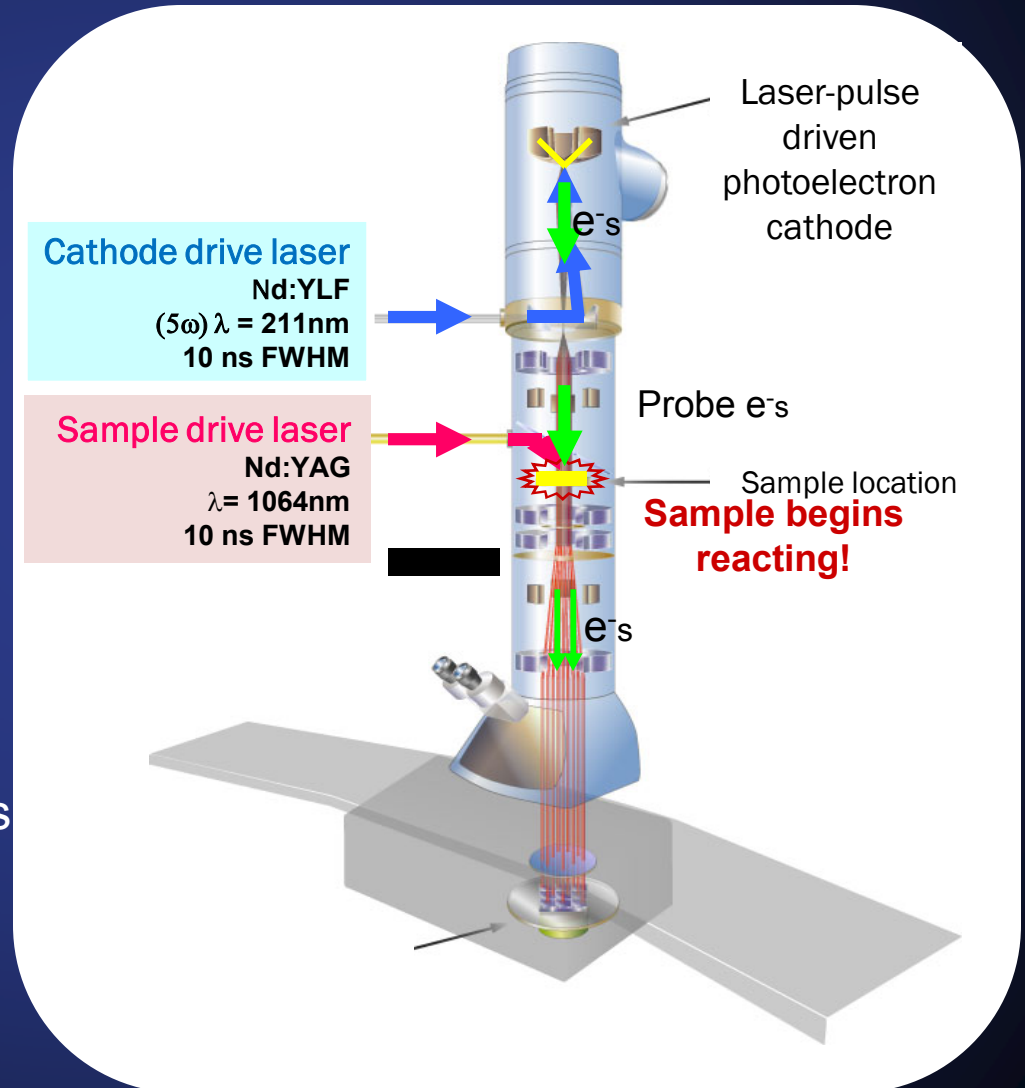
Space-time from a materials point of view



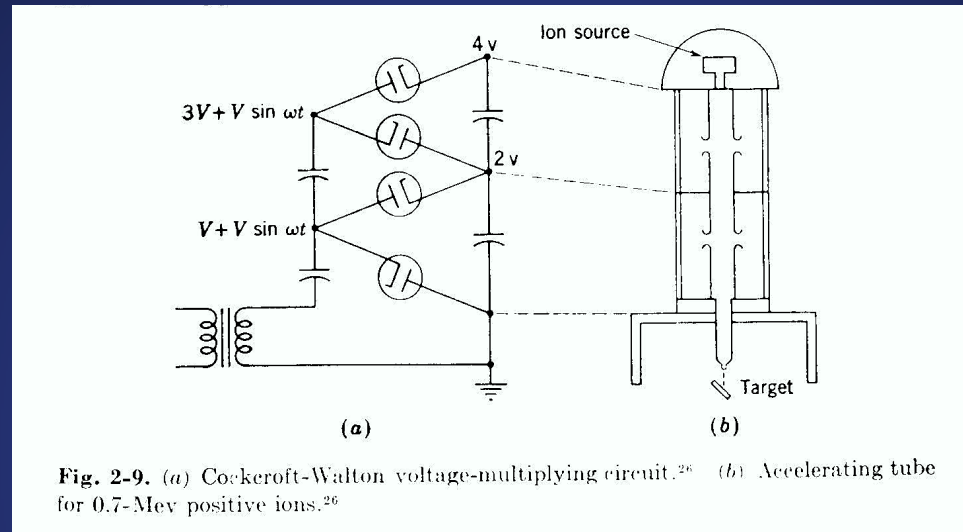
# DTEM: combining short electron pulses with traditional electron microscopy optics



- Started with a used JEOL 2000FX
- Standard thermionic filament replaced by high-brightness photocathode
- Access provided for photocathode laser
- Access provided for drive laser
- Low-noise, Peltier-cooled CCD camera installed
- LLNL brings unique skills in high-power lasers and associated optics



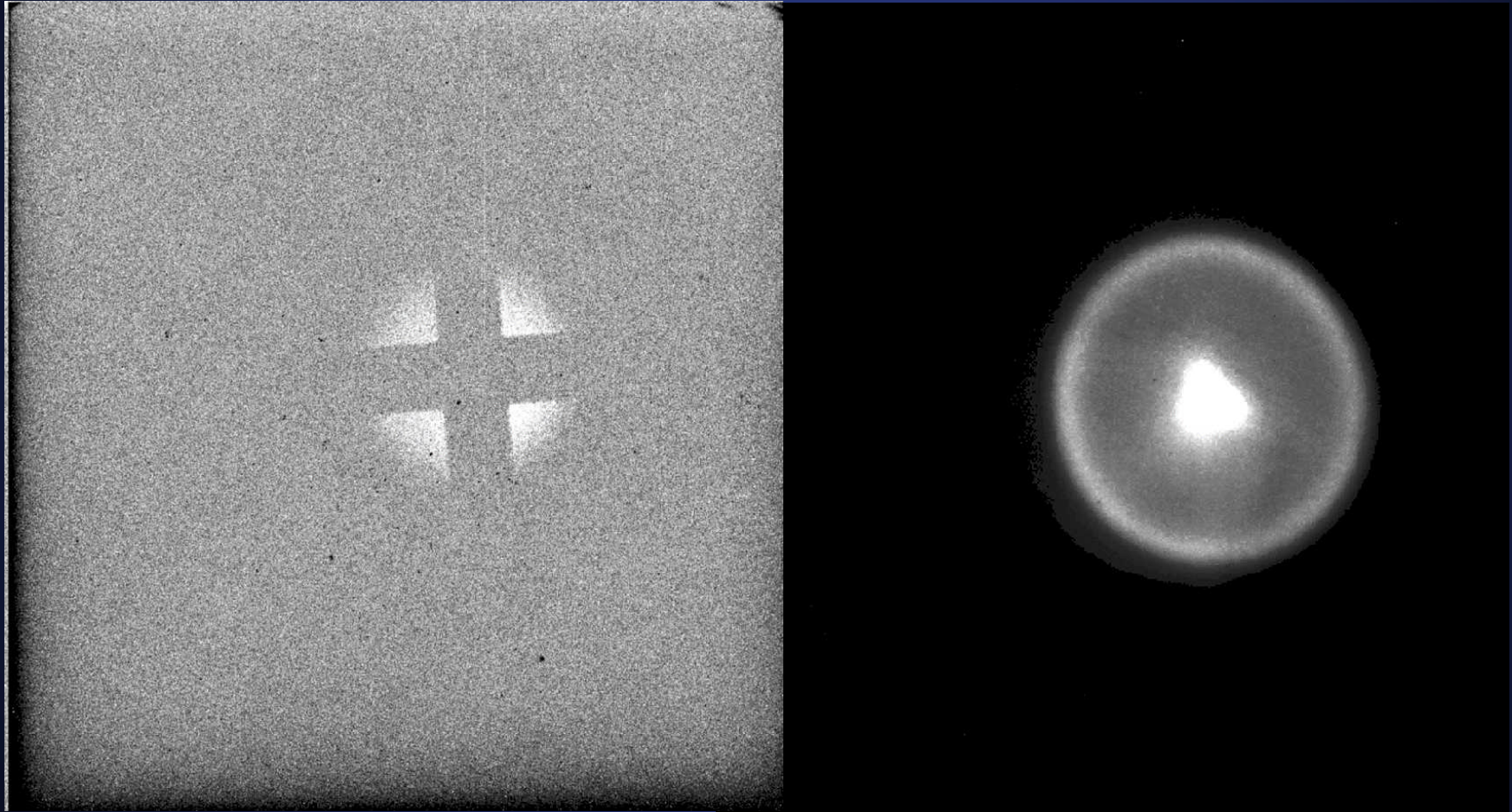
# The High Voltage electron source was a Cockcroft-Walton voltage multiplier



*Particle Accelerators, Livingston & Blewett, 1962*

- Frequency and number of sections had been optimized for the design CW current. What about pulsed?
- Intended 10ns pulses  $\ll 1/f \sim 100\mu s$
- 100pF of RG-220 style cable for  $RC \sim 20ms$
- Expected droop of 1 ppm;  $< 10ppm$  of cw ripple
- No further modifications needed for single-shot microscopy

# First high-quality pulsed images were obtained in January of 2005





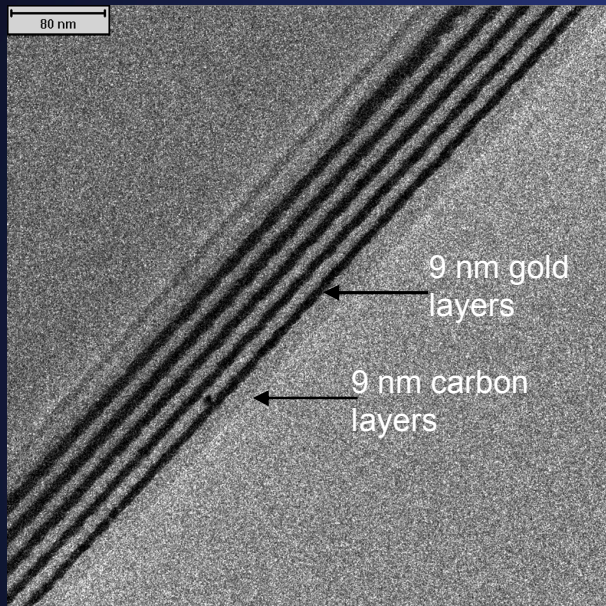
# First diffraction images were taken in March, 2005



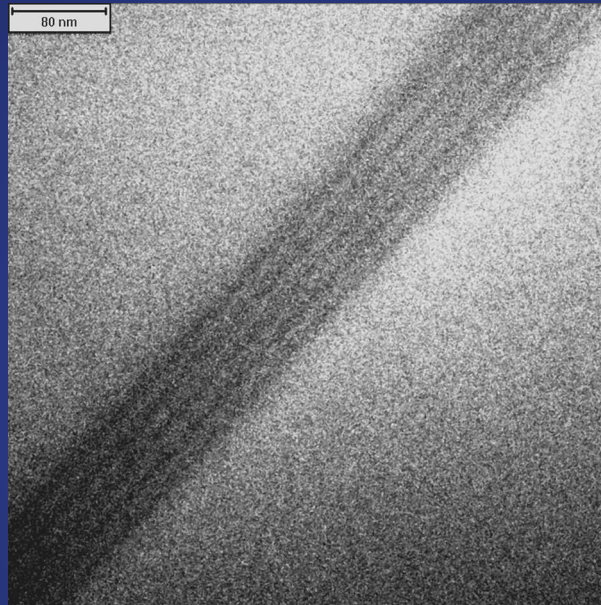
- Diffraction at a small aperture leads to these Airy rings
- Analogous to single slit diffraction pattern extended to radial geometry
- We were on our way!



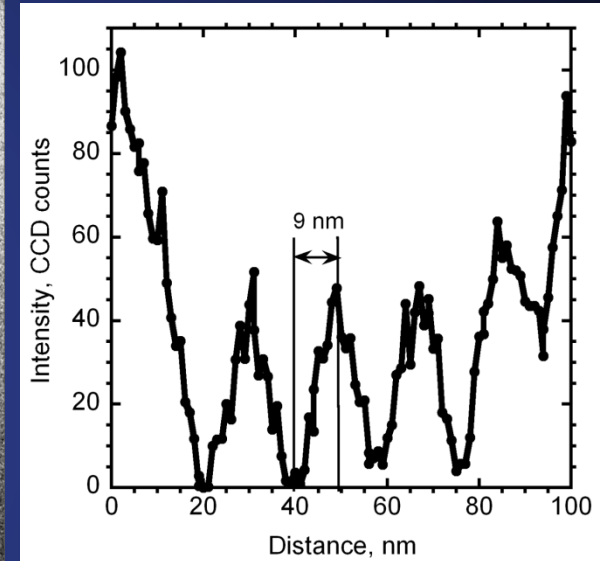
# The DTEM can resolve $<10$ nm features in a 15 ns exposure.



Conventional TEM Image



Single-Shot 15 ns DTEM Image



Intensity Trace

*Experiments by Tom LaGrange*



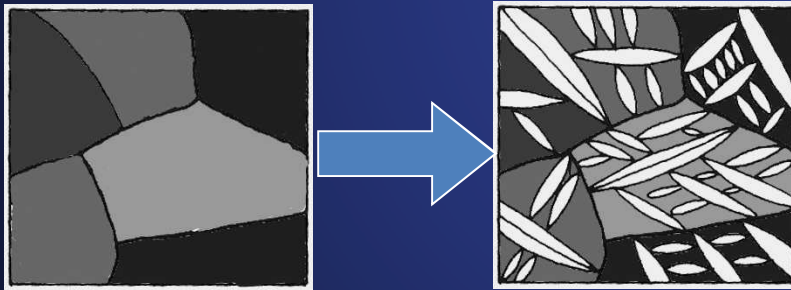
# So what good is this?

- Show me a phase transition!

# The $\alpha$ to $\beta$ transition in Ti is a martensitic transformation at high heating rates



- Martensitic transformations are fast
  - Often thought to proceed at the speed of sound
- Occurs upon heating to above 1155K (882 °C)
- Transforms from  $\alpha$  HCP to  $\beta$  BCC



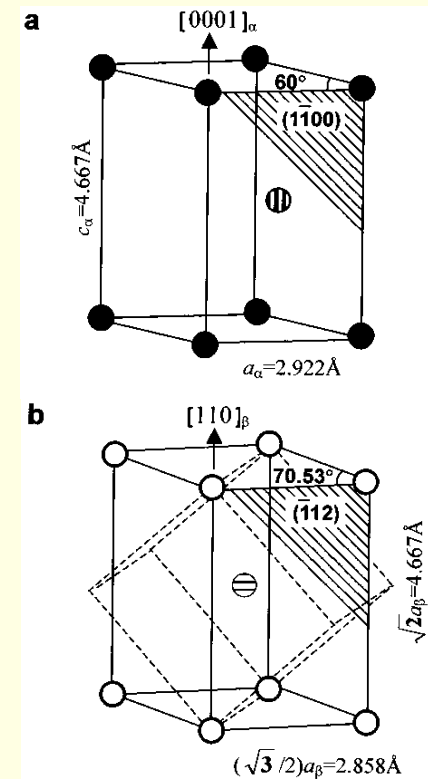
Experiments by Tom LaGrange

## The transformation shears the crystal

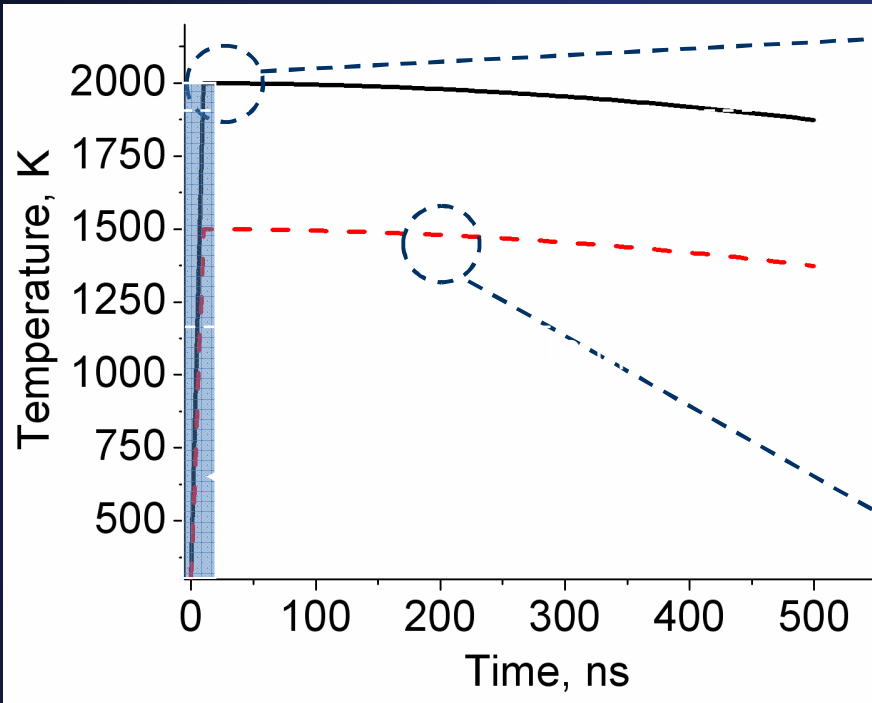
HCP



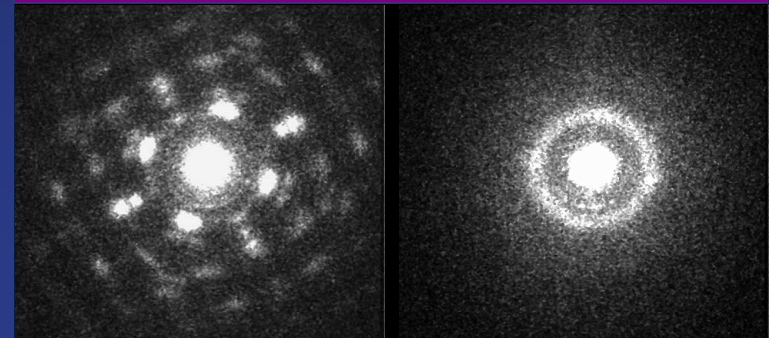
BCC



# Noting the threshold laser energy for melting calibrates film temperature in terms of laser energy



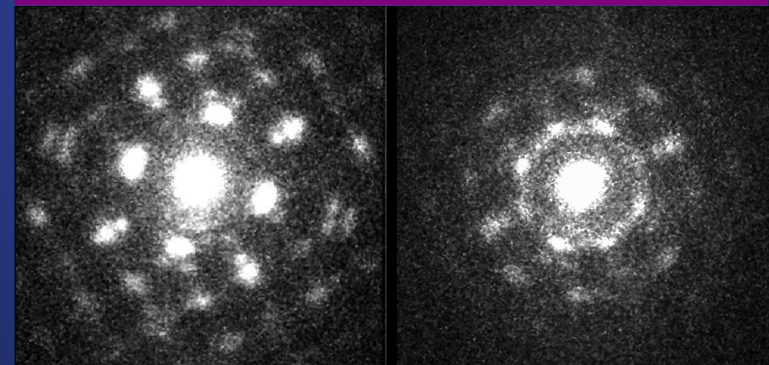
Melting transition



hcp Ti,  $\alpha$ -phase

Melt structure

Phase transformation ( $\alpha$  to  $\beta$ )



bcc Ti,  $\beta$ -phase

$$\alpha E_{in} = n(C_p \Delta T + \Delta H_{tr} + \Delta H_f)$$

T. LaGrange, et al., *Journal of Material Science* 41 (2006) 4440



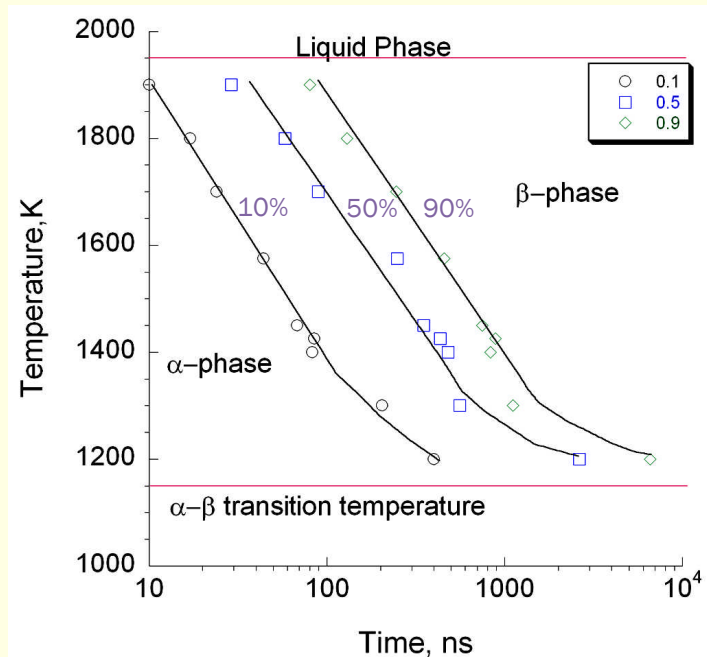
# Phase fraction data were used to create the first TTT diagram for nanocrystalline Ti films



- High temperature (near melt)
  - transformation is very rapid
- Low Temperatures
  - Transformation is (relatively) slow
- Compared to CALPHAD calculations
- Future experiments will assess the effect large grained materials on the transformation rates
- First Ti Time Temperature Transformation curve published

These data cannot be obtained in any other way

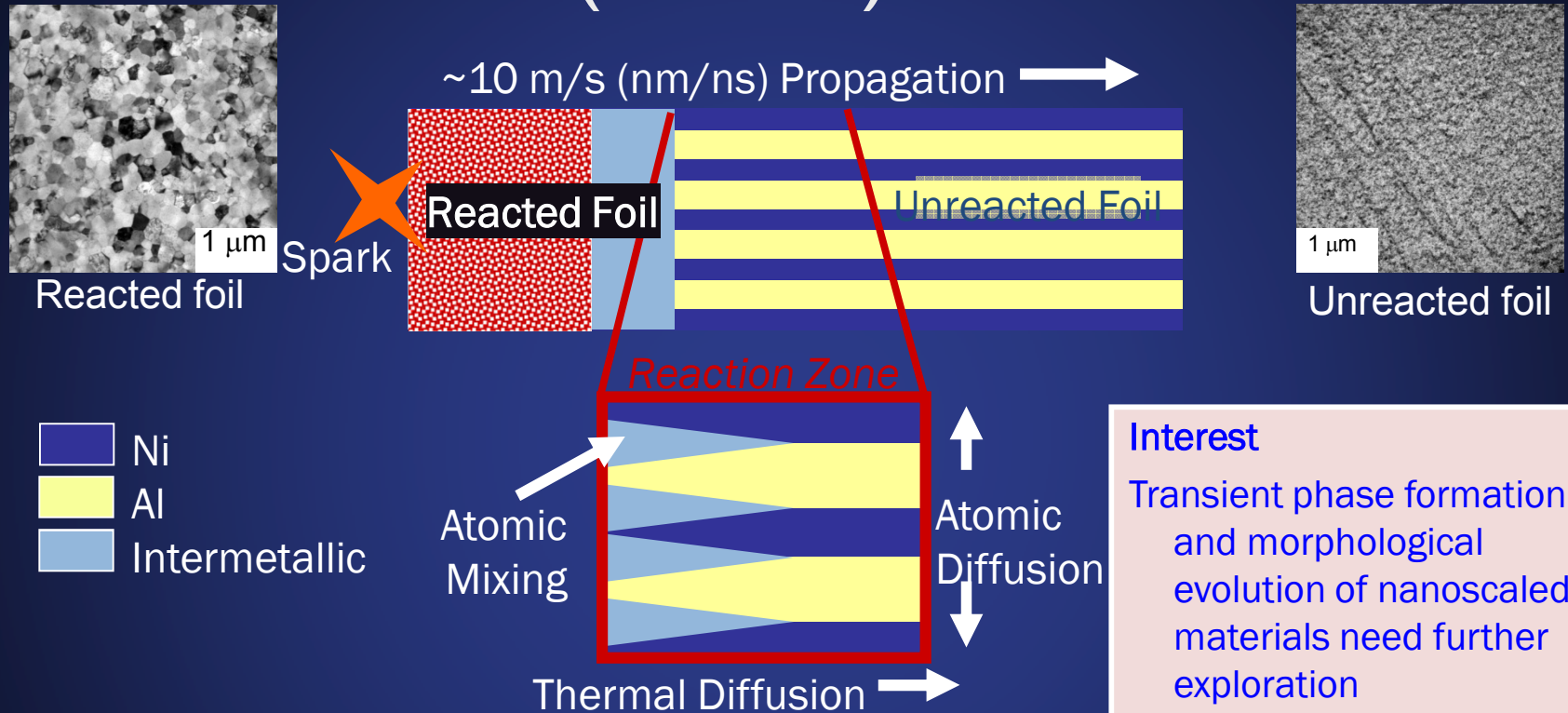
## First TTT isothermal diagram for Ti



A compilation of over 1000 DTEM experiments

T. LaGrange, et al., *submitted to Acta Materialia*

# Dynamic Reactive Multilayer Foils (RMLFs)



1. Thin transition metal bi-layers mix together
2. Exothermically form intermetallics
3. Self-propagating reaction of new phases travels across foil at high speeds

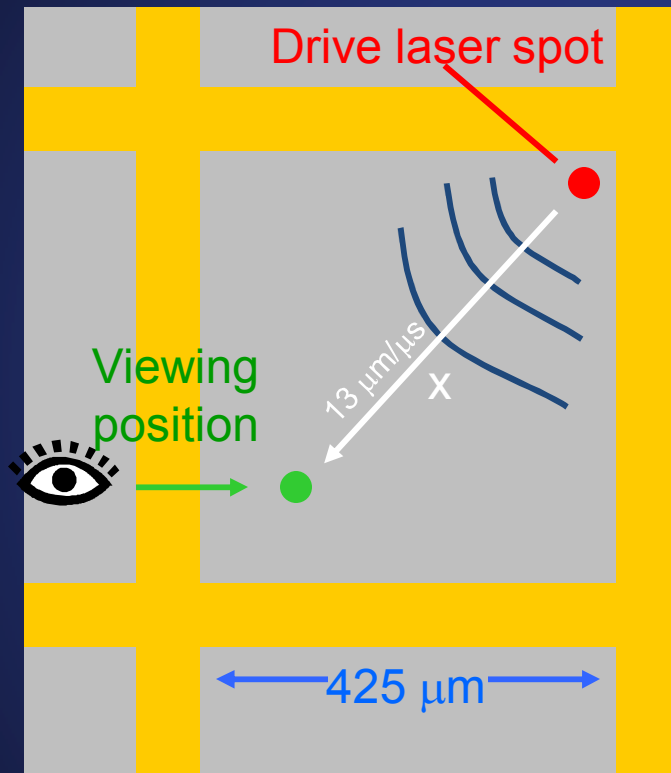
## Interest

Transient phase formation and morphological evolution of nanoscaled materials need further exploration

Direct metastable state observations cannot be done conventionally

Past studies have only used electron microscopy on quenched RMLFs

In RMLF experiments, the steady state reaction is initiated 100s of  $\mu\text{m}$  away from the field of view.



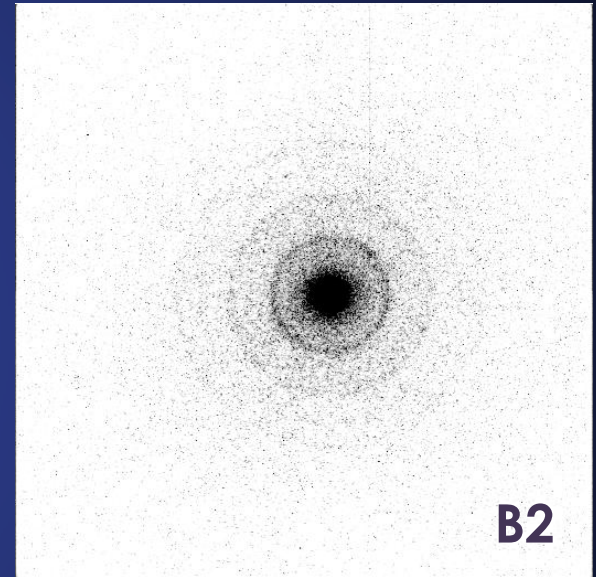
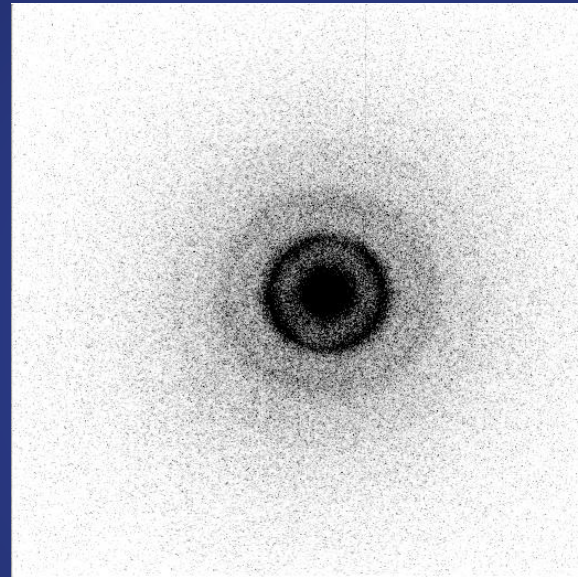
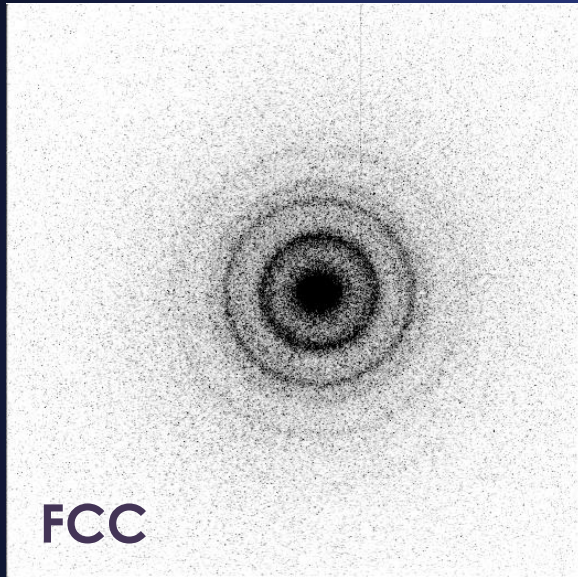
Cu TEM grid with RMLF sample

- Steady state reaction front speed is 13 m/s
- Experiment is completely contained within one grid square
- Large grid size minimizes thermally conductive effects of Cu
- Distance  $x$  can be calculated from low magnification TEM images

# Time-Resolved RMLF Experimental Data



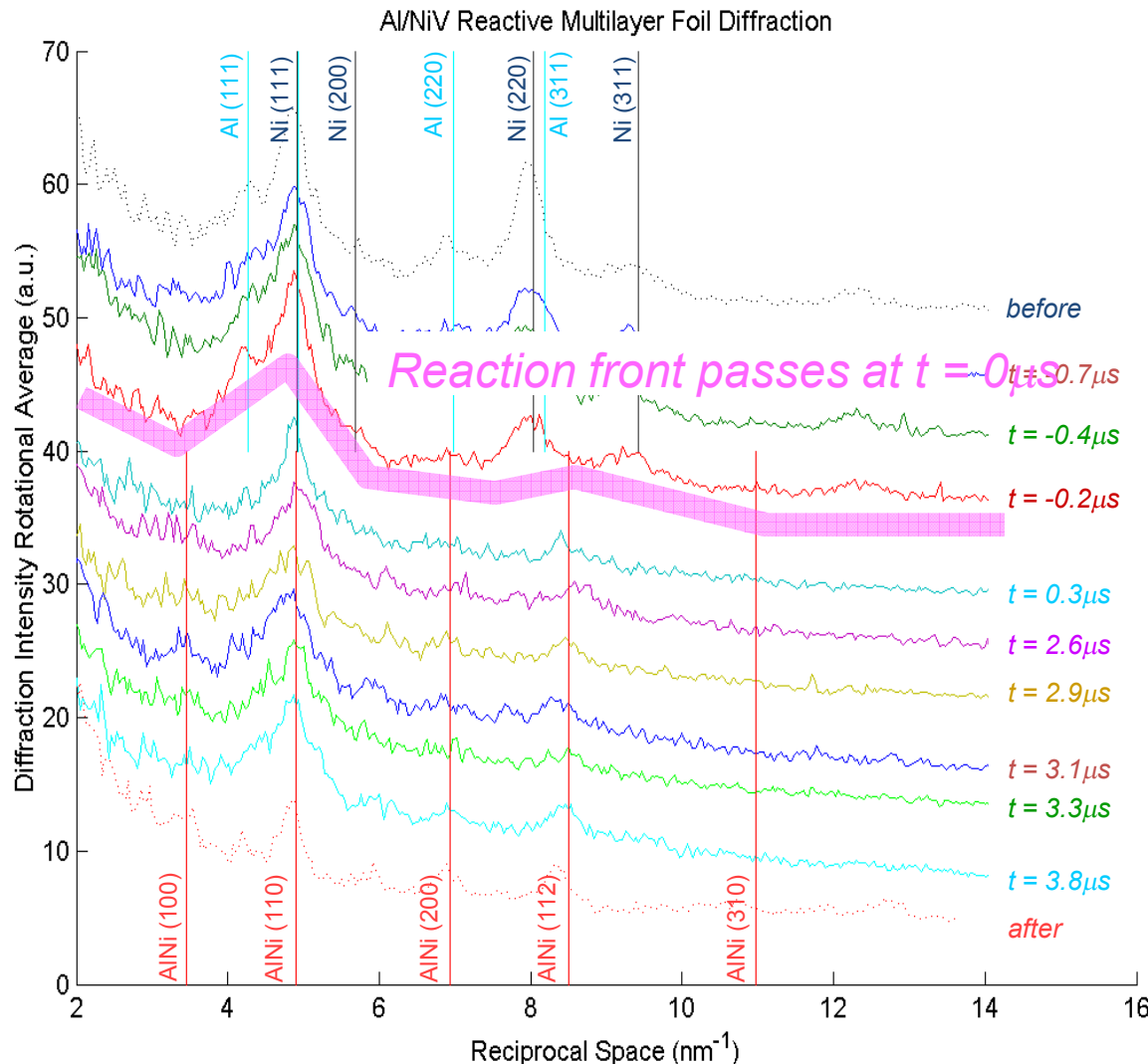
“Snap-shot” diffraction patterns with 30 ns time-resolution



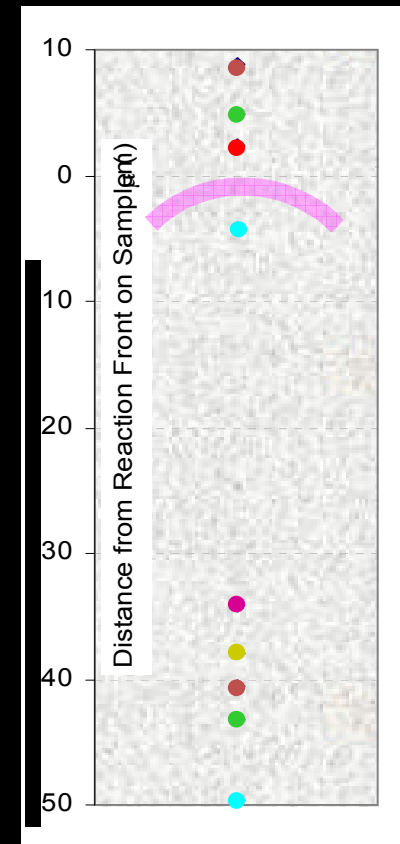
Ring spacing shows phases present in the sample as they vary through time.



# Phase Formation Timing is Found with Time-resolved Diffraction



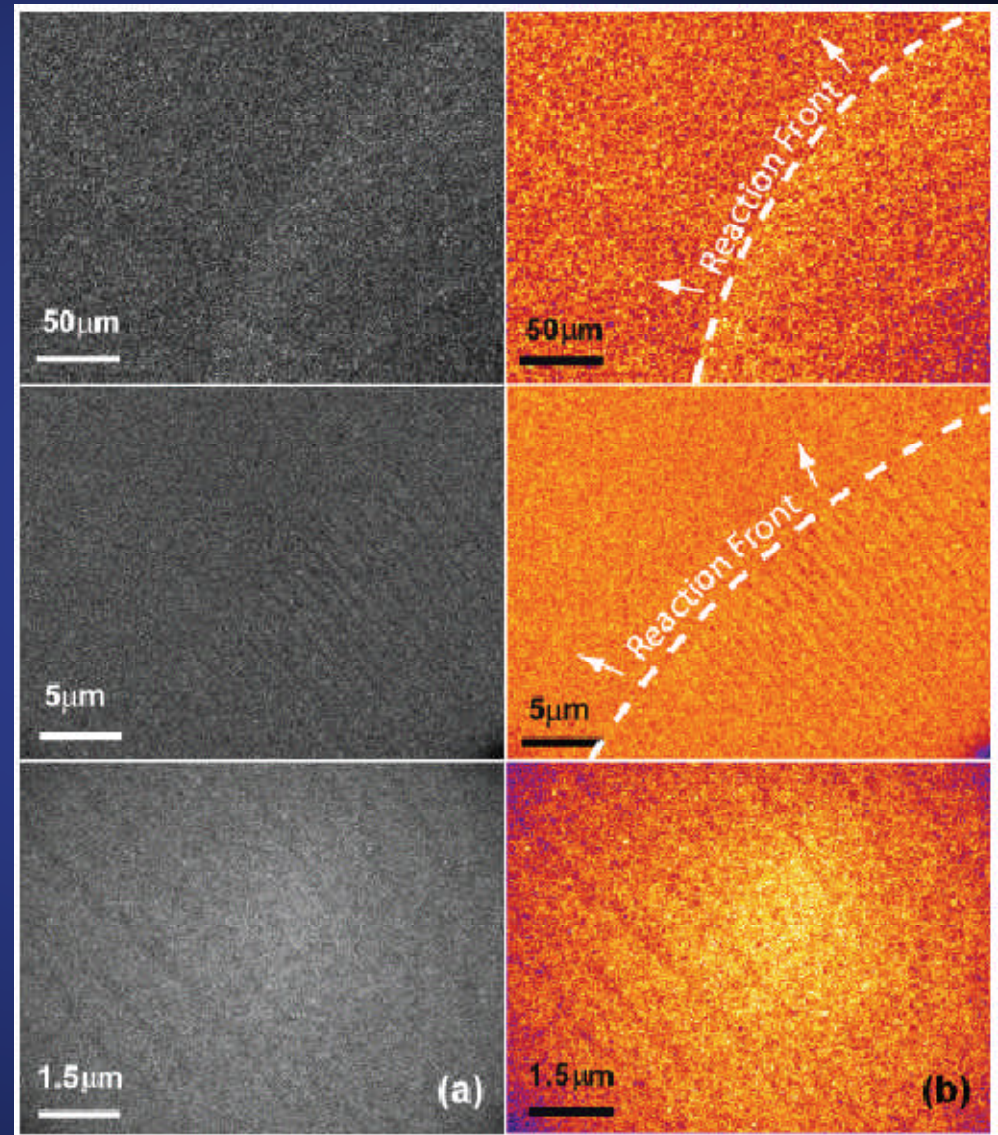
- ✓ Initial AlNi phase formation is detected within a 500ns gap
- ✓ AlNi is formed only 100's of ns after the reaction front has passed
- ✓ What is the reaction front morphology?



# Reaction Front Images reveal Transient Structure



- Fast plan-view TEM images acquired with 30ns resolution
- Reaction front is imaged to reveal microstructural details for the first time
- Dark lines appear 670nm apart
  - Appears to be cellular phase formation



(a) Raw data, (b) 3x3 median filter, false coloring.

Experiments by Judy Kim

# What is wrong with these studies?



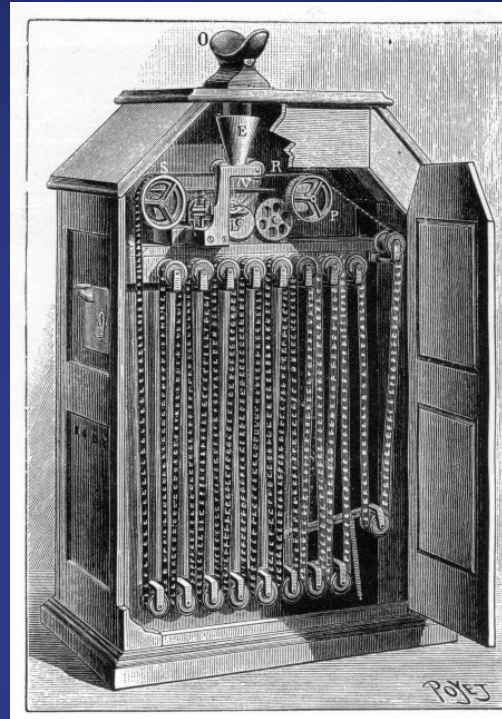
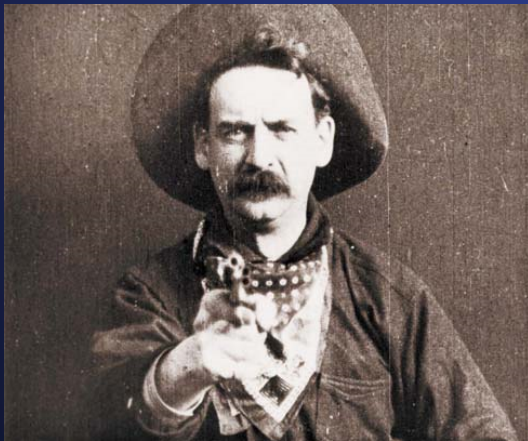
- Each “shot” involves a separate pump and probe pulse.
- To take a series, we change the timing of pump and probe
- This requires looking at a new piece of sample
- We assume pump pulses are repeatable and samples are uniform
- Better to have one pump pulse and multiple probe pulses, i.e. a movie.



# Everyone loves the movies



8-legged boar, 18,000BC, Spain



Edison's Kinetoscope,  
1889-1909, 25 cents



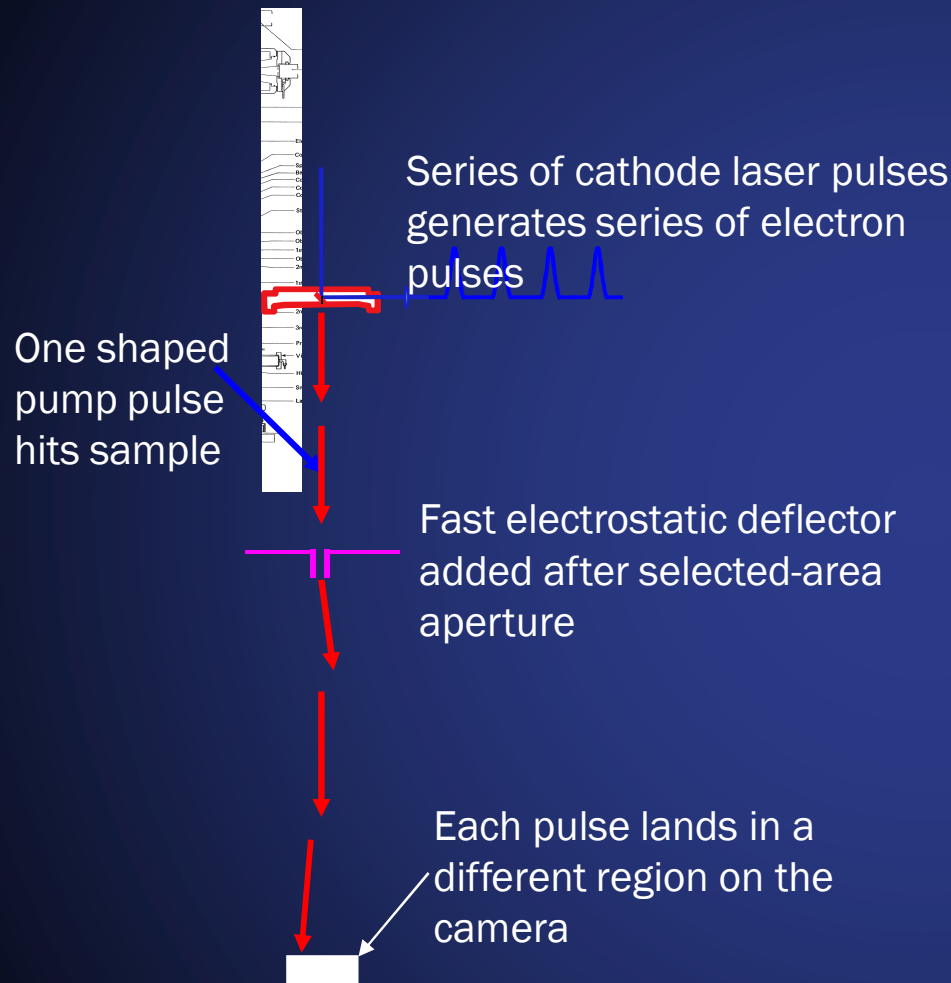


# What are the requirements for MM-DTEM?

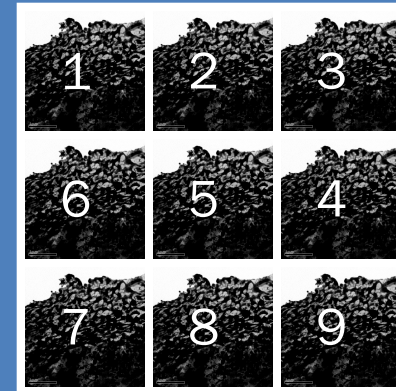


- Maintain 2-20 ns temporal resolution
- Variable frame rate of 15ns to 10's of us
- At least 9 frames total over a 1cm CCD
- Deflect images/patterns by 1cm over 60cm
  - 16 mrad deflection angle

# Movie Mode lets us capture a series of images or diffraction patterns from a single drive pulse



Read out entire CCD at end of experiment and segment into frames:



## Key Issues

- Deflected beams must maintain beam quality
  - Aberrations must be controlled or correctable
  - Below the projector lens
- Deflection hardware must be vacuum-relevant and manufacturable
  - 100um beams in 1 mm structures
  - Arc-free
- Deflection electronics must be close to off-the-shelf

## Previous demonstrations:

- Takaoka and Ura, 1983, multiple frames, ms switching
- Bostanjoglo, ca. 1990, 2-3 frames, 20 ns switching



# Magnetic or Electrostatic?

- Magnetic dipole
  - 200 kV means  $\beta \sim .7$ ,  $v = 20$  cm/ns
  - 8 mrad means 13 gauss deflection field
  - 13 G over 1mm x 1cm means 10 amps
  - Single-turn coils mean 12 nH inductance
  - $L di/dt$  in 10ns means 10 volts
  - But a microscope is full of iron!
- Electric dipole
  - Max gradient of 100V over 1mm
  - 1cm axial extent means 30 ps in the transverse field
  - Lorentz force  $eE = ma$  means  $v_{\text{transverse}} \sim 1\text{m/us}$
  - 3mm deflection over 60 cm throw

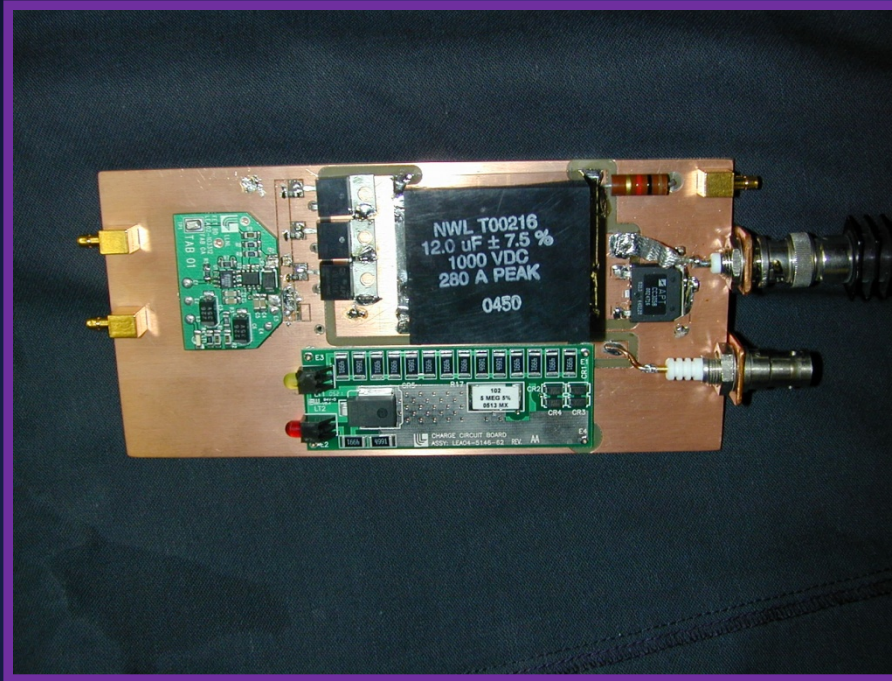
# Quadrupole field switching



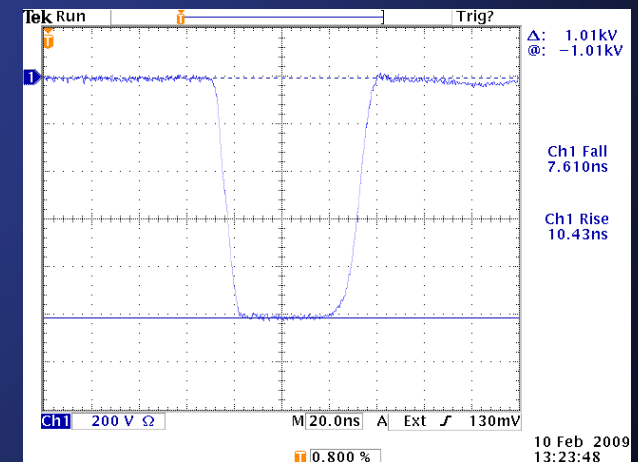
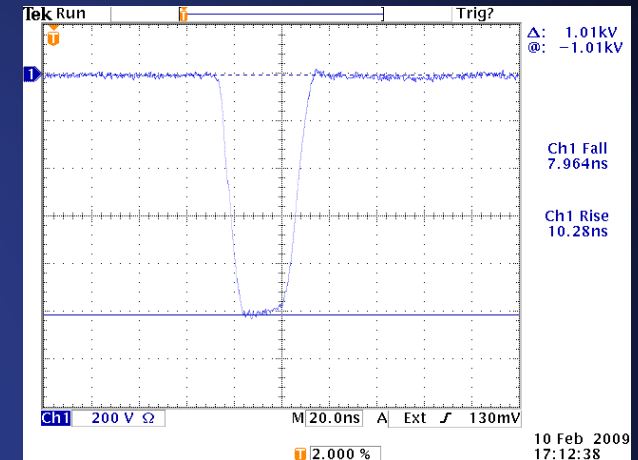
|   |   |   |  |   |   |   |   |
|---|---|---|--|---|---|---|---|
|   | + |   |  | + |   | + |   |
| + |   | - |  | 0 | 0 | - | + |
|   | - |   |  |   | - |   | - |
|   | 0 |   |  | 0 |   | 0 |   |
| + |   | - |  | 0 | 0 | - | + |
|   | 0 |   |  | 0 |   | 0 |   |
|   | - |   |  | - |   | - |   |
| + |   | - |  | 0 | 0 | - | + |
|   | + |   |  | + |   | + |   |

- RF amps
  - Agile
  - Need slew rate  
10V/ns
- Series of caps and closing switches
- Beam interception and charging a possibility

# MOSFET-based pulser boards will drive electrostatic deflector plates

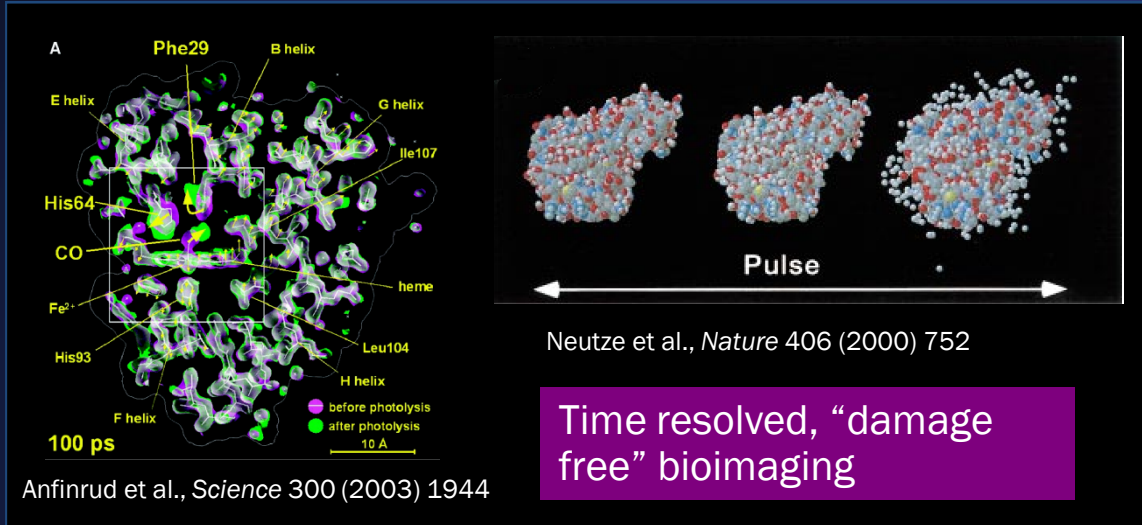


- Using 3 ea APT 4M120K MOSFETs. These devices are fast-switching and rated for 1200V, 15A pulsed.
- Using existing gate drive circuit developed for DARHT
- Using existing charge/discharge circuit developed for NIF
- 12 $\mu$ F, 1kV Capacitor used for NIF

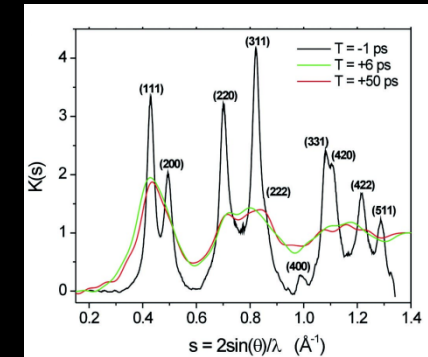




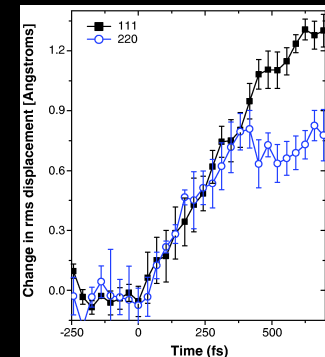
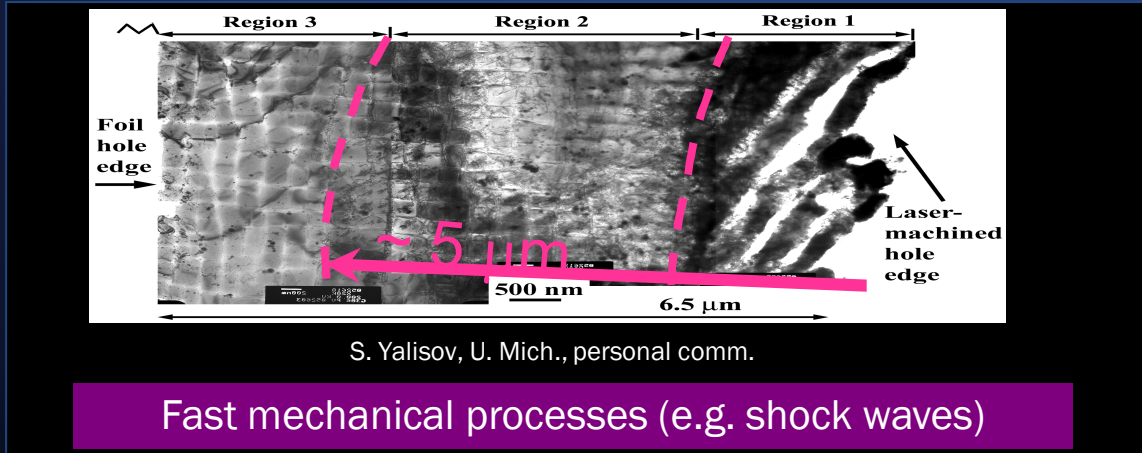
# Ultrafast, high resolution imaging offers great scientific dividends



## Non-thermal processes



Siwick et al., *Science* 302 (2003) 1382

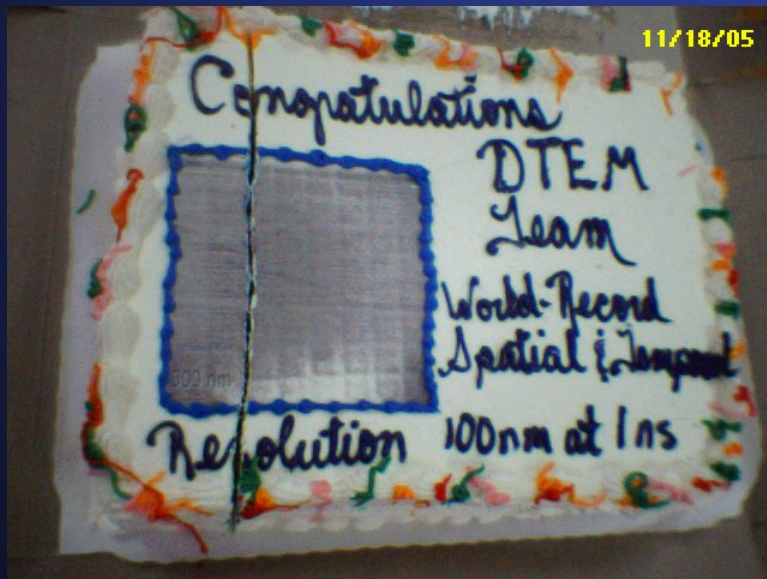


Lindenberg et al., *Science* 308 (2005) 392

# Cheers to the DTEM team for a great device and a bright future in the movies!



- Chemistry: Wayne King, Geoff Campbell, Bryan Reed, Tom LaGrange, Nigel D. Browning
- Global Security : Brent Stuart
- Engineering: Ed Cook, Bill DeHope, Ben Pyke, Rich Shuttlesworth



**2005: First Images**



**2008: R&D-100 Awardees**

